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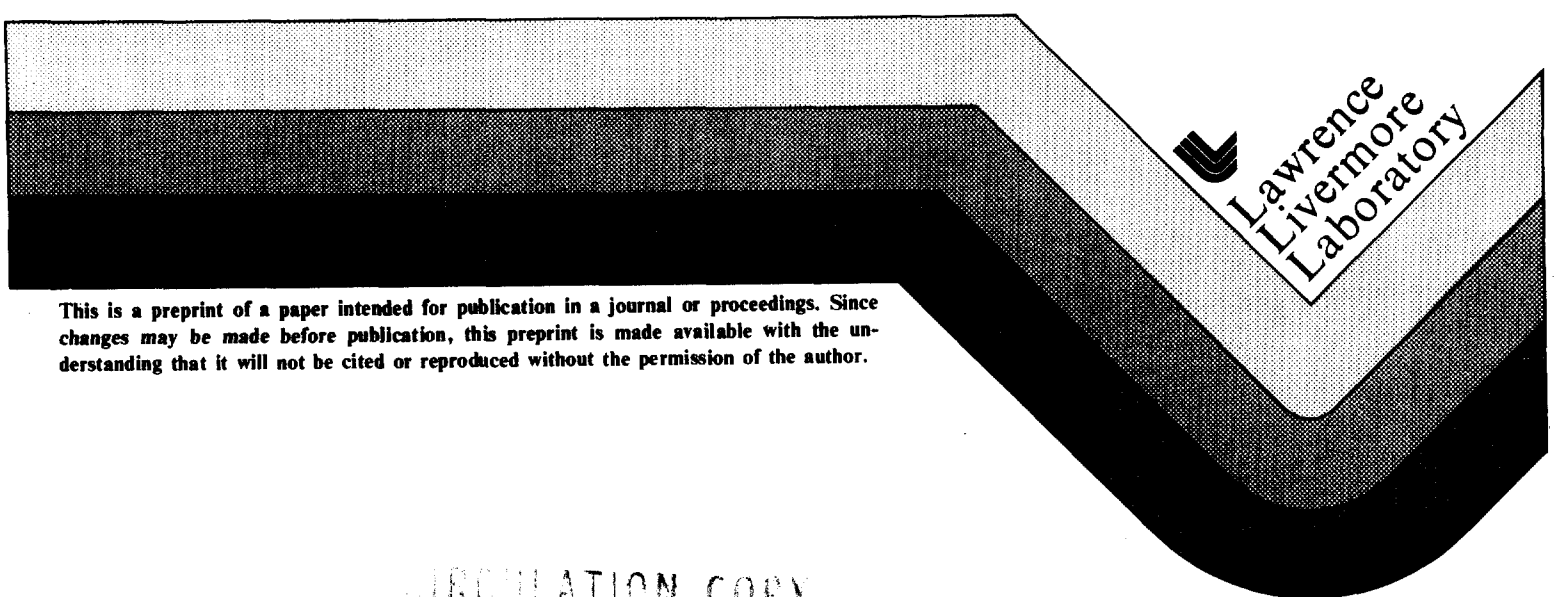

FORMATION OF H^- AND D^- IONS BY PARTICLE BACKSCATTERING FROM ALKALI/TRANSITION METAL COMPLEXES*

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Abstract

The data for negative ion reflection yields is analyzed using a backscattering model for the secondary emission coefficient. The enhancement of the secondary emission coefficient is discussed in terms of reflection, formation, and survival probabilities. The yield of negative ions from alkali/transition metal surfaces by low energy atoms emitted from the ion-source discharge is calculated. Volume production of negative ions generated by plasma-surface interactions in a low-work-function-surface bucket-discharge is discussed.

I. Introduction

The central theme of this paper is the generation of negative ions by hydrogen particles reflecting from surfaces. The early observations of negative ions produced by hydrogen molecular ions incident on nickel surfaces have been reviewed by Massey.[1] New evidence is accumulating for surface-ion production in ion-source discharges,[2,3] evidence which is supported by a growing body of surface backscattering data.[4,5,6] On the astrophysical scale, the negative ion composition of planetary and lunar ionospheres is thought to be sustained by continuous particle bombardment and reflection from the planetary surfaces.[7,8] The presence of negative ions in cometary atmospheres may also have their origin in particle backscattering processes.[9] It would seem that the full scope of these different phenomena is still only lightly perceived, and reflection yields may provide a new paradigm for a variety of diverse and novel phenomena.

We shall be concerned with the formation of H^- and D^- as caused by hydrogen and deuterium particles backscattering from alkali metals and alkali/transition metal complexes. The main thrust of this presentation is directed towards the explication and enhancement of negative ion yields from ion source plasma-surface interactions. The range of incident particle energies of interest here extends from the threshold incident energy, E_{TH}, given by the difference of the surface work function and the negative ion affinity,[10] $E_{TH} = \phi - A$, upwards to incident energies of order one keV. The experimental data on reflection yields is used in conjunction with a theoretical model for the negative-ion-secondary-emission-coefficient, NISEC, which is used both for the interpretation of the data and as a basis for extrapolating the negative ion yields down to incident energies near the threshold region. In this paper the terms reflection and backscattering are used interchangeably.

II. NISEC

Our model for NISEC is taken to be the product of the reflected particle velocity distribution, the reflected angular distribution, the formation probability for negative ions in the near-surface region, and the survival probability of negative ions as they move to great distances away from the surface.[11-13] The functional form for the formation and survival probabilities assumes the underlying granular structure of the crystal surface can be replaced with a uniform charge distribution, in analogy with the jellium surface model,[14] and that these probabilities can be taken to be functions only of the perpendicular (normal) component of the backscattered particle velocity, $v_\perp = v \cos\theta$. The NISEC for a particle with incident energy E_i is then

$$\mathrm{NISEC}(E_i) = 2 \iint f_i(v) \cos\theta \left[1 - e^{-\frac{\alpha}{v\cos\theta}}\right] e^{-\frac{\beta}{v\cos\theta}} \, dv \, d(\cos\theta) \, . \qquad (1)$$

This expression is integrated over $\cos\theta$ to yield a single integral over the backscattered velocity,

$$\mathrm{NISEC}(E_i) = 2 \int f_i(v) \, g(\alpha, \beta, v) dv \, . \qquad (2)$$

The reflected particle velocity distributions, $f_i(v)$, are generated using the Marlowe Monte Carlo reflection code developed at Oak Ridge by the Solid State Physics Group.[15,16]

The experimental data is analyzed by inserting experimental NISEC values on the left

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hand side of Eq. (2) for several different values of incident energy, E_i, and adjusting the α, β to obtain a least-squares fit to the data. The semi-emperical values for α, β found in this way then determine the formation and survival probabilities, $1 - \exp - \alpha/v \cos\theta$, and $\exp-\beta/v \cos\theta$, respectively, that enter into the integrand of Eq. (1).

In Figure one is shown the NISEC data[6] for protons incident upon the alkalies Li, Na, K, Rb, and Cs plotted as a function of incident proton energy. The lower case letters superimposed on the data indicate the least squares fits of Eq. (1) to the experimental data for the respective alkalies. In Figure two is shown a similar set of data but for incident deuterium.

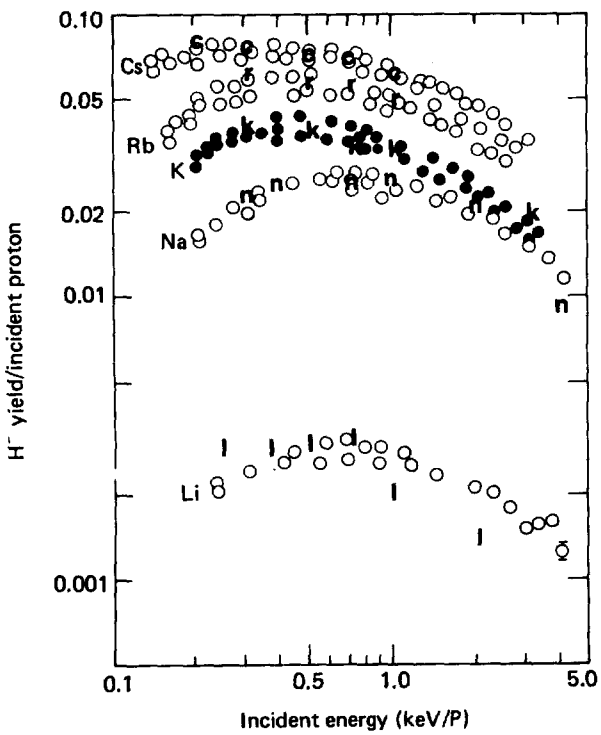

Fig. 1. NISEC vs incident proton energy.

Figure three summarizes the formation and survival probabilities plotted here as a function of the hydrogen perpendicular energy; the α, β that enter into these probabilities are taken from the least squares fits of the first two figures. The formation probabilities approach unity at low energy and decrease monotonically toward higher energies, while the survival probabilities increase toward higher energies. Note that at any particular energy the survival probability varies in an inverse way with work function, i.e., these probabilities increase from lithium through cesium. (The lithium curve has been suppressed but would lie immediately below the sodium curve.) The dashed lines are the survival probabilities for potassium and cesium calculated using a resonant electron transfer model and reported at the previous Symposium in this series.[17] The

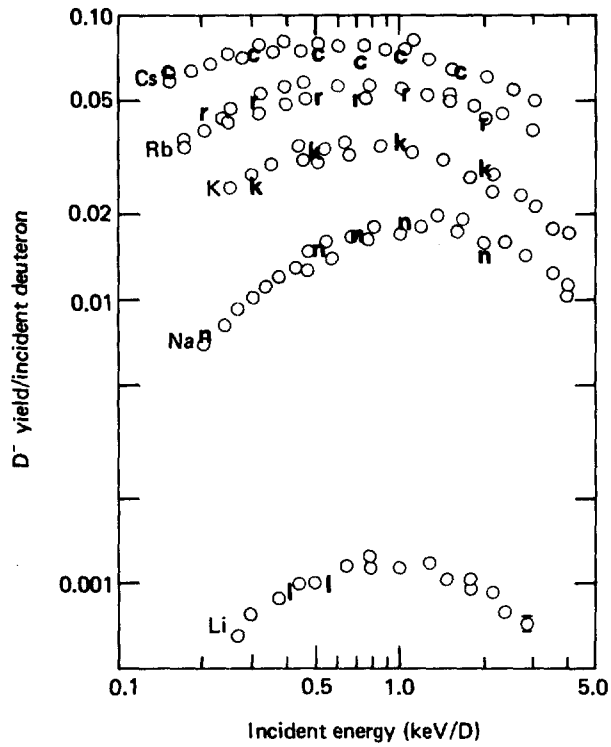

Fig. 2. NISEC vs incident deuteron energy.

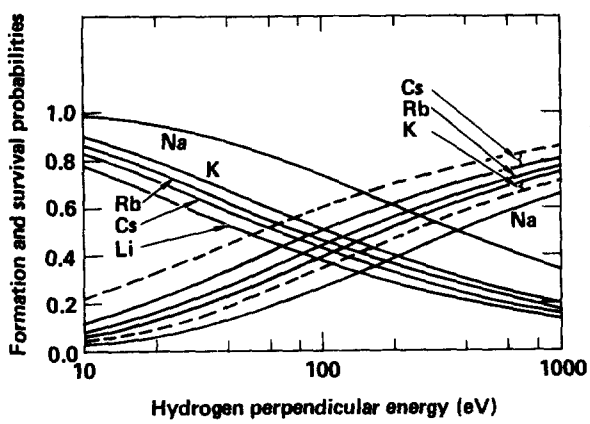

Fig. 3. Formation and survival probabilities for the alkalies.

agreement between the calculated and semi-emperical values is not precise but is sufficiently close to suggest that no essential effects have been neglected. Figure one, two, and three are taken from reference 13.

III. NISEC Enchancement

Although the alkali targets provide relatively large yields, these yields can be enhanced by reflection from alkali/transition metal complexes for which the adsorbate alkali is a partial monolayer coating chosen to give a

minimum surface work function, and the substrate chosen to give high particle reflectivity. We shall consider the NISEC to be approximated by three factors: particle reflection, formation, and survival probabilities.

The enhancement of the particle reflection coefficent for normally incident deuterium particles is discussed in reference 11. In this paper it is shown that for low-Z substrates the reflection coefficient is an increasing of function of atomic number Z up to some intermediate value, and remains roughly constant for higher Z values. For 300 eV incident deuterium the reflection coefficient varies only slightly for elements with atomic number greater than that of cesium. For lower energy 50 eV deuterium, the coefficient increases rapidly with Z, reaching an approximately asymptotic value for a nickel substrate. For nickel or higher Z substrates, the optimum substrate material can be selected independently of particle reflection considerations.

The underlying surface phenomena affecting the magnitude of the formation probability are still only partly understood. In reference 13 we present evidence to the effect that the density of occupied electronic states near the Fermi level of the surface is a relevant parameter. We have already noted that a general feature of the formation probability is to increase towards unity as the perpendicular energy component of the backscattered particles is decreased.

The survival probability at low energies is especially susceptible to enhancement by working with partial-monolayer alkali/transition metal complexes. The electric dipole layer that is formed between the adsorbate layer and the substrate inhibits the tunneling loss of the electron from the outwardly moving negative ion to the metal substrate; if the adsorbate layer is non-conducting, the survival probability that results can remain near unity for energies as low as a few electron volts.[17]

In figure four is shown the NISEC versus incident ion energy for protons incident on Cs/Cu and deuterons incident on Cs/Ni together with the deuterium-cesium data of figure two. The deuterium least-squares fits are indicated by slanted crosses, the hydrogen fits by vertical crosses. In the case of the composite layers the cesium coverage was varied to give the optimum negative ion yield shown here. Inspection of the figure shows a considerable increase in NISEC for the composite surfaces compared with cesium. Note in particular that the yield for Cs/Ni is increasing at the lower energies. Our projection using Eq. (1) shows this NISEC reaching a maximum for energies below 100 eV.

In figure five is a comparison of the survival probabilities derived from the least-squares fits of figure four. A substantial improvement in survival probability occurs at lower energies for the composite systems, but still short of the limiting theoretical expectation for a non-conducting adsorbate.

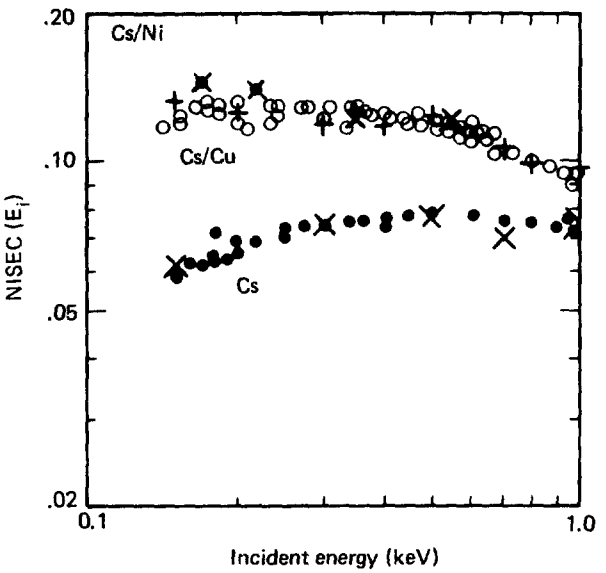

Fig. 4.

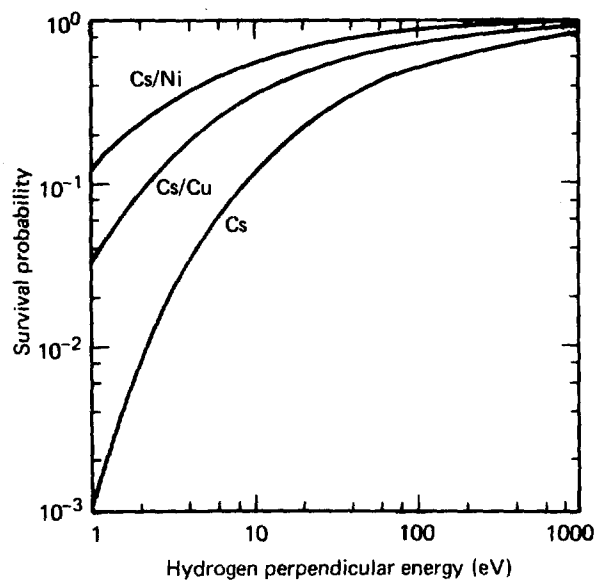

Fig. 5.

The survival probability can presumably be enhanced further by increasing the strength of the adsorbate/substrate electric dipole layer while maintaining a low minimum work function, ϕ_m. The strength of this layer is related to the change in work function, $\Delta\phi$, that is equal to the difference of the minimum composite work function, ϕ_m, and the clean substrate work function, ϕ_s. A semi-emperical relation for $\Delta\phi$ for alkali/transition metal complexes has been given by Swanson and Strayer,[18]

$$\Delta\phi = k_1(k_2 I_a - \phi_s) \quad .$$

Here k_1 and k_2 are emperically determined constants, and I_a is the ionization potential of the isolated adsorbate atom. For a particular adsorbate, e.g., cesium, $\Delta\phi$ has its largest (negative) magnitude for substrate materials with the largest work function. In Table I are listed the optimum cesium/transition metal complexes in the order of decreasing $|\Delta\phi|$ for both polycrystalline and monocrystalline substrates. The ϕ_s values are taken from the paper of Michaelson.[19] The ϕ_s and $\Delta\phi$ are in units of electron volts.

Table I

Polycrystalline				Monocrystalline		
	ϕ_s	$\lvert\Delta\phi\rvert$		Crystal Face	ϕ_s	$\lvert\Delta\phi\rvert$
Cs/Pt	5.65	4.21	Cs/Ir	111	5.76	4.33
Cs/Ir	5.27	3.79		100	5.67	4.23
				110	5.42	3.96
Cs/Ni	5.15	3.66	Cs/Re	1011	5.75	4.32
Cs/Pd	5.12	3.63	Cs/Pt	111	5.7	4.26
Cs/Au	5.1	3.61	Cs/Pd	111	5.6	4.15
Cs/Co	5.0	3.50	Cs/Au	100	5.47	4.01
Cs/Rh	4.98	3.48	Cs/Ni	111	5.35	3.88
				100	5.22	3.74
Cs/Re	4.96	3.46		110	5.04	3.54
Cs/Te	4.95	3.44	Cs/W	110	5.25	3.77
				100	4.63	3.10
Cs/Os	4.83	3.31		111	4.47	2.92
Cs/Ru	4.71	3.18	Cs/Mo	110	4.95	3.44
Cs/Cu	4.65	3.12		111	4.55	3.01
Cs/Mo	4.6	3.06		100	4.53	2.99
Cs/W	4.55	3.01	Cs/Cu	111	4.94	3.43

The polycrystalline values for ϕ_s listed here must be used with caution since these values can vary as much as 0.3 eV depending on how the substrate material is prepared. Selenium exhibits the largest known work function, 5.9 eV, but is not a transition element and is not listed here because its cesium adsorbtion properties are unknown.

In the table we have referred only to cesium adsorbates because cesium will provide the lowest values for ϕ_m. If we were selecting the adsorbate/substrate combination on the basis of $\Delta\phi$ alone, other alkali adsorbates would be interpolated among the entries.

Inspection of the table suggests that a significant increase in the survival probability over that of polycrystalline Cs/Ni may be possible, particularly for certain monocrystalline substrate crystals of Ir, Re, and Pt.

Finally, several workers have confirmed a low work function, $\phi_m = 1.1$ eV, and a large $|\Delta\phi| = 4.1$ eV for a Cs/O/W complex.[18,20,21] By further saturating with oxygen a maximum $|\Delta\phi| = 6.4$ eV can be achieved, but at the cost of increasing ϕ_m to 1.4 eV. However, the presence of oxygen in the O/W substrate is expected to reduce its reflectivity; whether or not the improved values for ϕ_m, $\Delta\phi$ can offset this reduced reflectivity in the overall NISEC is uncertain.

IV. The Fast Atom Flux

Given the prospect of relatively large survival probabilities in the low energy, 1 - 10 eV range, we consider next the backscattering yield of negative ions due to energetic atoms emanating from the discharge plasma. Negative ion production by few electron-volt atoms incident on a cesium-coated surface has been reported by Graham.[22] A low energy peak in the distribution of negative ions is reported at this Symposium in the paper by Leung and Ehlers. Some plasma and wall reactions that produce energetic atoms with energies above one electron volt are

$$e + H_2 \longrightarrow H(m\ell) + H(n\ell) + e \qquad (a)$$

$$e + H_2 \longrightarrow H(n\ell) + H^+ + 2e, \qquad (b)$$

$$H_2^+ + H_2 \longrightarrow H_3^+ + H, \qquad (c)$$

$$H_2^+ + e(\text{wall}) \longrightarrow H + H \qquad (d)$$

$$H_3^+ + e(\text{wall}) \longrightarrow H_2 + H, \qquad (e)$$

$$H_3^+ + e(\text{wall}) \longrightarrow 3H \qquad (f)$$

$$H_3^+ + H^- \longrightarrow H_2 + 2H, \qquad (g)$$

$$H_3^+ + H \longrightarrow 4H \qquad (h)$$

$$H_2^+ + H^- \longrightarrow 3H. \qquad (i)$$

The corresponding deuterium reaction rates would be quite similar to the hydrogen rates since these reactions are essentially all electronic processes.

Another source of very energetic atoms arises from the slowing down of positive ions initially accelerated through the cathode or convertor sheath and then successively reflected off the discharge walls. The particle current density of these neutrals may be of the same order-of-magnitude as the positive ion current density striking the cathode,[10,23,24] with an energy distribution extending downward from the incident positive atomic energy. The negative ions resulting from this atomic flux are not considered in this section.

Process (a) for $n = m = \ell = 1$ proceeds through the first excited $b\,{}^3\Sigma_u^+$ electronic state of the molecule[25] yielding atoms with kinetic energies of 2 - 4 eV and peaked near 2.7 eV. For $n = m = 2$ process (a) yields[26] excited atoms with kinetic energies of approximately 3.8 eV. Excitation for $n = 1$, m, leading to energetic atoms in the 1 - 12 eV

range have also been identified.[27] Reaction (b) proceeds mainly through the H_2^+ first excited $^2\Sigma_u^+$ state yielding an equal-energy proton-atom pair with the energy distribution of either particle peaked near 8 eV.[28,29] Process (c) yields an atom whose energy distribution is dependent upon the initial vibrational level of the ions on the right and left. The final vibrational distribution of H_3^+ is uncertain, but the atom energies will range from zero up to 3.25 eV. The corresponding H_2 channels in reactions (d) and (f), and reaction (e) have already been discussed as a means for producing vibrationally excited molecules in a discharge.[30]

In figure six is shown the fast atom energy distributions obtained for processes (a) and (b). The 2.7 distribution has been calculated using an $H_2(v = 0)$ ground state vibrational function[31] and a delta-function final state. The curves labeled 50 eV and 75 eV are measured proton distributions obtained by energetic electron collisions via process (b). Momentum and energy conservation imply similar distributions for the atomic fragments. The backscattered distribution of H^- ions obtained from the fast atom flux will be distorted from the incident distribution due to energy transfers to the crystal lattice and to the energy dependence of the survival probability. For 10 eV hydrogen particles incident on nickel or tungsten Marlowe shows the backscattered distributions peaked near 9 eV. Also shown in the figure for comparison is the backscattered distribution of H^- ions obtained using Eq. (1) for 200 eV hydrogen ions incident on cesium.

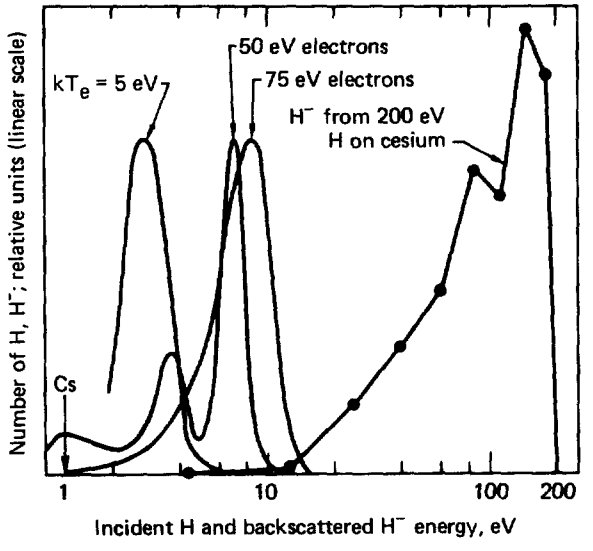

Fig. 6.

The rate of growth of the atom density in the discharge due to reaction (a) is given by

$$\frac{dn_H}{dt} = 2n_o n_e \langle\sigma v\rangle - \frac{n_H}{b}\frac{V}{L} \quad , \qquad (4)$$

where n_o and n_e are the gas and electron densities, V/L the atom collision rate with the walls, and b the number of atom wall bounces prior to H^- formation or sticking.

The fast atoms are formed in an isotropic distribution, and those with glancing collisions will have small survival probabilities. We can underestimate the yield by considering only the fraction incident within a 90° cone centered on the normal, i.e., one-eighth of the total distribution, and take the mean perpendicular energy to be one-half the incident energy for purposes of estimating the survival probability. Using the equilibrium atom density obtained from Eq. (4), we calculate the negative ion current density equal to the product of the NISEC, N, and the atomic flux onto the active connector electrode to be

$$j_- = \frac{N}{4} n_o n_e \langle\sigma v\rangle bL \quad . \qquad (5)$$

To estimate the current density we shall choose the following parameters appropriate to a high-power discharge[3]:

$kT_e = 5$ eV

$n_o = 8 \times 10^{13}$ molecules cm^{-3}

$n_e = 1 \times 10^{12}$ electrons cm^{-3}

$L = 10$ cm

$f_1 = .2$

$f_2 = .4$

$f_3 = .4$

$\overline{\sigma v}(a) = 0.5 \times 10^{-8}$

$\overline{\sigma v}(b) = 0.18 \times 10^{-8}$

$\overline{\sigma v}(c) = 0.2 \times 10^{-9}$

$b \leq 3$ such that $Nb \leq 1$

Using these parameters and Eq. (5) or its equivalent for reactions (b) and (c) we have calculated the negative ion current densities emitted from the convertor electrode for different surfaces and listed these current densities in Table II. The NISEC's have been evaluated using reflected fractions 0.55 and 0.9 for Cs and Ni, respectively,[32] and survival probabilities taken from figure four. Included is the limiting case for a survival probability near unity and NISEC = .9. For reaction (c) we have used the same NISEC's as for reaction (b). The table entries are listed in units of mA cm^{-2}.

Reaction	Cs	Cs/Ni	NISEC = .9
(a)	.75	70	160
(b)	.09	.6	1.3
(c)	.8	13.	13.

Inspection of the table shows that interesting current densities may be obtained via reaction (a). For any particular reaction it is evident that the negative ion yield will

be very sensitive to the condition of the surface coverage.

Reactions (d), (e), and (f) may provide a flux of energetic atoms larger than the equivalent ion flux onto the convertor cathode, depending on the relative area of the discharge chamber and the convertor cathode.

V. Volume Production of H^-, D^- Ions Via a Plasma Surface Cascade

In this section we shall consider the volume production of negative ions in a discharge that is confined by a magnetic bucket system with walls coated to provide a low work function, large $|\Delta\phi|$ surface. The electric potential between the plasma and wall is presumed to be a minimum to allow negative ions formed by wall reflection to join the plasma via bucket collisions and ion-ion coulomb-collisional energy deposition. The plasma is presumed to be sufficiently dense that negative ion collision rates are dominated by ion-ion collisions:

$$D^- + D^+ \longrightarrow 2D \quad , \qquad \text{(i)}$$

$$D^- + D_2^+ \longrightarrow 3D \quad , \qquad \text{(ii)}$$

$$D^- + D_3^+ \longrightarrow 4D \quad . \qquad \text{(iii)}$$

Reactions (ii) and (iii) proceed through the D_2 triplet channels, and for lack of detailed information we shall assume these couplings occur in the ratio of 3 to 1 compared with the singlet channels. Interaction potentials for reactions (i) and (ii) are available,[33,34] and (ii) will be taken as the prototype for (iii).

The first step in reaction (ii) may proceed via either of the two triplet states

$$D^- + D_2^+ \longrightarrow D_2(a^3\textstyle\sum_g^+,\ c^3\Pi_u) + D(1s) \quad , \qquad \text{(iv)}$$

and will release about 4.8 eV, 3.2 eV of which appears as D(1s) kinetic energy. The triplets formed will ultimately decay to the $D_2(b^3\Sigma_u^+)$ repulsive state in times of order 10^{-8} to 10^{-5} seconds, releasing two D(1s) atoms with kinetic energies ranging up to about 3.5 eV.

If f_1, f_2, f_3 are the respective species fractions of the positive ions, $\overline{\sigma v}(+,-)$ is the reaction rate taken equal for each of the above reactions, the rate of growth of the atom density in the discharge is given by

$$\frac{dn_D}{dt} = n_-\left[2 + \frac{1}{4} f_2 + f_3\right] n_+ \ \overline{\sigma v}(+,-) - \frac{n_D}{b}\frac{V}{L} \quad , \qquad \text{(v)}$$

where V/L is the collision rate of the atoms with the walls and b is the number of times an atom will collide with the wall before sticking to the wall or being converted to a negative ion. If G is the fraction of wall area activated for negative ion formation, N the NISEC for fast atom collisions, and $\overline{\sigma v}$ the electron collisional detachment rate, the rate of growth of the negative ion density in the discharge by negative ions returning from the wall becomes

$$\frac{dn_-}{dt} = GN\frac{V}{L} n_D - n_-\left[n_+ \ \overline{\sigma v}(+,-) + n_e \ \overline{\sigma v}\right] \quad . \qquad \text{(vi)}$$

The rate Eqs. (v) and (vi) are coupled through n_-, n_D. It is easily shown that the growth of the negative ion density given by (vi) will increase exponentially provided that

$$\left[GNb\left(2 + \frac{1}{4} f_2 + f_3\right) - 1\right]$$

$$n_+ \ \overline{\sigma v}(+,-) - n_e \ \overline{\sigma v} > 1 \quad . \qquad \text{(vii)}$$

If $kT_e \leq 3$ eV and $n_e \leq n_+$, then $n_+ \ \overline{\sigma v}(+,-) \geq n_e \ \overline{\sigma v}$. In the case where the equalities hold the exponentiation conditions are the most stringent and the term in the brackets must be greater than unity. Taking as an example $f_1 = .1$, $f_2 = .4$, $f_3 = .5$, and noting that $Nb \leq$ unity, exponentiation occurs for $G > .77$ provided Nb is unity. If kT_e is less than 3 eV or n_e is less than n_+, these conditions are relaxed.

In circumstances where the exponentiation condition is met, fast atoms produced in ion-ion collisions strike the active walls and return as negative ions which in turn generate a new and larger generation of fast atoms, etc. This cascading processes will continue until the negative ion density approaches the positive ion density, at which point the cascade is interrupted by a changing plasma potential.

VI. Acknowledgments

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